

C.3 MARKETS

The leading consumers of refined copper are wire mills, accounting for 75% of the refined copper consumption. Brass mills producing copper and copper alloy semi-fabricated shapes are the other dominant consumers at 23%. The dominant end-users of copper and copper alloys are the construction and electronic products industries, accounting for 65% of copper end-usage. Transportation equipment, such as vehicle radiators, accounts for an end-usage of 11.6%. A passenger car typically contains 50 lb of copper wire (BHP 1997). Copper and copper alloy powders are used for brake linings and bands, bushings, instruments, and filters in the automotive and aerospace industries, for electrical and electronic applications, for anti-fouling paints and coatings, and for various chemical and medical purposes. Copper chemicals, principally CuSO_4 , CuO , and Cu_2O , are widely used in algacides, fungicides, wood preservatives, copper plating, pigments, electronic applications, and numerous special applications.

End-use markets for brass rod include:

- construction and remodeling 48%
- industrial equipment and machinery 30%
- electrical and electronics 8%
- transportation equipment 8%
- exports 4%
- consumer durables 2%

Typical products include plumbing fixtures, industrial valves and fittings, welding and cutting equipment, cable and electronic connectors, gas grill components, brake hose assemblies, and decorative hardware.

C.3.1 Scrap Prices

Scrap prices are related to the refined copper price, but the price spread must be sufficient to allow for collection, sorting, shipping, chopping, etc. If the price spread is too narrow, the processor cannot charge enough for the end product, which also is determined by the refined copper price, to make a profit. When refined copper prices are high, more copper scrap is offered to processors. If refined copper prices are low, less scrap enters the market. As the gap between

scrap price and refined price narrows, the processing cost may make the scrap uneconomical (Carlin et al. 1995).

C.3.2 Scrap Consumption

Copper-base scrap consumption in 1997 by type of scrap and by processor is summarized in Table C-11 (Edelstein 1998). The total consumption of 1,755,000 t is greater than the total of 1,370,000 t shown in Table C-4 because the latter table is based on the copper content of the scrap while the former is based on the gross weight of the copper-base alloys. Both of these tables are based on copper-base scrap, while Table C-3 includes other alloys where copper is not the primary alloying element. Table C-11 emphasizes the diversity of copper scrap uses. Unalloyed scrap is consumed by smelters, refiners, ingot makers, brass mills, wire-rod mills, and foundries. While about 63% of alloy scrap is consumed by brass mills, a significant fraction is also processed by ingot makers, smelters, refiners, and foundries.

It is worth noting that environmental restrictions on lead associated with copper pose obstacles to recycling certain copper alloys, particularly some brasses. The addition of up to 8% lead in brass castings and rod improves machinability and casting characteristics. New drinking water standards may require elimination of most of the lead from brass plumbing fixtures (Carlin et al. 1995). As can be seen in Table C-11, leaded brass is a major component of copper-base scrap recycling.

C.4 PARTITIONING OF CONTAMINANTS

This section discusses the manner in which impurities partition during the various metallurgical operations involved in the refining of copper scrap.

The main application of copper is as an electrical conductor. As such, extremely high purity levels are required to maintain low electrical resistance. As little as 0.08% iron or 0.05% phosphorus will reduce the conductivity of copper by 33% (CDA 1998b). Typical output from the cathode furnace may be electrolytic tough-pitch copper which contains a minimum 99.90% copper or oxygen-free copper, which contains a minimum of 99.95% copper. Thus, the aim of copper refining is to remove most of the impurities from the metal. The following sections discuss the expected distribution of contaminants in scrap that is introduced into the copper processing cycle (see Figure C-2). The expected partitioning from scrap which is introduced into brass mills, foundries, and the like will be discussed in a later section.

C.4.1 Partitioning During Copper Refining

C.4.1.1 Thermochemical Considerations

Most impurities in copper scrap introduced into blast furnaces, converters, or anode (fire refining) furnaces will tend to be oxidized during processing and removed with the slag. Theoretically, this will include all oxides whose free energies of formation per gram-atom of oxygen are more negative than that of CuO. The free energy of formation of CuO at 1,500 K (1,227°C) is about -6 Kcal/gram-atom of oxygen (Glassner 1957). Oxides of metals such as Po, Te, and the platinum group (Pt, Pd, Rh, Ir) are less stable than CuO and the respective metals should remain with the copper. Cs₂O boils below 1,000 K and would be volatilized. Other species with low boiling points such as Cd, Po, Ra, Se, and Zn may also be partially volatilized (see Table E-3). Relevant free energy data for various oxides are summarized in Table C-12. Of the elements whose oxides are listed in this table, only Ag and Ru are expected to remain in the copper under equilibrium conditions.

Copeland et al. (1978) calculated the partition ratios between copper and an oxide slag for several contaminants, based on free-energy data. The authors assumed that: (1) the weight of the slag was 10% of the weight of the metal, (2) the activity of the copper oxide in the slag was 0.1, and (3) the activity of the contaminant oxide in the slag was 0.01. Henry's Law constants for the contaminant and the contaminant oxide were assumed to be unity (i.e., ideal solution behavior). The partition ratio was defined as the weight of the contaminant in the slag divided by the weight of the contaminant in the ingot. Calculated partition ratios at 1,400 K are summarized in Table C-13. These calculations suggest that all the elements listed except cobalt will partition to the slag and that concentrations of most of these contaminants in the copper will be very low.

However, blister copper leaving the converter is reported to contain small amounts of impurities such as As, Bi, Fe, Ni, Pb, Sb, Se, Te, and precious metals (Davenport 1986). This emphasizes that predictions based on thermochemical calculations and vapor pressures are only guidelines to impurity behavior during processing.

C.4.1.2 Experimental Partitioning Studies

Some experimental work has been done to measure partitioning of radionuclides during copper smelting. Heshmatpour et al. (1983) found that plutonium strongly partitioned to the slag, as would be expected from thermodynamic considerations. Three tests were conducted, in which

500 ppm of PuO_2 was melted with 200 grams of copper in recrystallized alumina crucibles at $1,400^\circ\text{C}$. The slag weight was 10% of the metal weight. Slags included a borosilicate composition (80% SiO_2 , 13% B_2O_3 , 4% Na_2O , 2% Al_2O_3 , 1% K_2O), a blast furnace composition (40% CaO , 30% SiO_2 , 10% Al_2O_3 , 15% Fe_2O_3 , 5% CaF_2) and a high silica composition (60% SiO_2 , 30% CaO , 10% Al_2O_3). The respective partition ratios (defined as the ratio of total Pu in the slag to total Pu in melt) were 3,225, 157, and 107. In each case less than 1 ppm of Pu remained in the copper. In the last two cases, a significant fraction of the input PuO_2 was not accounted for, rendering these values suspect.

Copeland and Heestand (1980) measured the partition ratio of uranium in copper in a laboratory experiment by equilibrating copper at $1,100^\circ\text{C}$ with a slag containing 0.3 wt% U. The measured partition ratio was 600, which is many orders of magnitude lower than the predicted value (see Table C-13). The final uranium concentration in the copper was 5 ppm. Other experimental details were not provided. A laboratory drip-melting experiment was also described, in which surface contaminated copper was placed on a screen and melted. The molten copper passed through the screen into a crucible below. Assay of the dross and the ingot showed that the former contained 3,400 ppm U, while the latter contained 1.4 ppm U. In a scaled-up experiment, about 40 kg of copper scrap surface contaminated with UO_2 was drip melted. The copper ingots contained 0.07 ppm U, while the slag contained 1,250 ppm U, resulting in a partition ratio of 18,000.

In subsequent work, Heshmatpour and Copeland (1981) conducted a series of laboratory experiments, in which 500 ppm UO_2 was added to small melts of copper produced with various fluxes. The samples were melted in recrystallized alumina or zirconia crucibles and held at about $1,250^\circ\text{C}$ to equilibrate the melt and the slag. The results, which are summarized in Table C-14, show that the partition ratios vary from 49 to 3182.

Mautz (1975) and Davis et al. (1957) summarized the results of melting 40 heats (about 100 tons) of uranium-contaminated copper scrap with surface activities up to 150,000 dpm/100 cm^2 in an oil-fired reverberatory furnace with a 125-ft stack. Ten samples taken from the copper product showed uranium values ranging from <0.022 ppm to 3.1 ppm. Six slag samples contained 1,440 to 1,730 ppm of U, while two samples contained only 0.43 and 0.47 ppm. No explanation for these low values was provided, although it is possible that the copper melts from which these slag samples were taken were initially very low in U. Uranium contamination of the furnace lining was also detected. Activity in the stack averaged 4×10^{-11} $\mu\text{Ci/cc}$. No air activity

was detected outside the furnace in excess of $1.7 \times 10^{-12} \mu\text{Ci}/\text{cm}^3$, which is 10% of the MPC value listed in NBS Handbook 52 for a controlled area. Samples collected to detect fallout showed no measurable uranium contamination of areas inside or outside the furnace building.

Table C-12. Standard Free Energies of Formation for Various Oxides at 1,500 K

Metal Oxide	$-\Delta F^\circ$ (Kcal/g-atom O)
Ag_2O	decomposes at 460 K
RuO_4	1.9
CuO	5.8
Cs_2O	9.4
Cu_2O	14.2
PbO	19.1
TcO_2	19.9
Sb_2O_3	26.0
CoO	26.5
NiO	26.5
FeO	38.6
ZnO	39.2
MnO	65.7
SiO_2	73.4
PaO_2	89.8
AmO_2	89.8
NpO_2	91.6
RaO	94.6
CeO_2	94.6
UO_2	99.0
Pu_2O_3	99.9
SrO	102
ThO_2	113

Source: Copeland et al. 1978

Abe et al. (1985) also conducted laboratory experiments to examine melt refining as a copper decontamination scheme. In these studies, 100 grams of metal and 10 grams of flux were melted in an alumina crucible under argon. Using a 1,550°C melting temperature, a melting time of one hour and a flux consisting of 40% SiO_2 , 40% CaO , and 20% Al_2O_3 , decontamination factors

ranged from 100 for an initial uranium concentration of 10 ppm to 10^4 for 1,000 ppm. The final uranium concentration in the ingot appeared to be relatively insensitive to the amount of uranium introduced into the melt. This suggests that the uranium content in the melt would not be less than about 0.1 ppm under the conditions of these experiments. However, the minimum observed uranium concentration in the melt-refined ingot—0.083 ppm—is very close to the 0.075 ppm of uranium in the copper feed stock used in this experiment.

Table C-13. Calculated Partition Ratios of Various Contaminants Between Copper and an Oxide Slag at 1,400 K

Contaminant	Partition Ratio
Th	10^{31}
Hf	10^{26}
U	10^{24}
Np	10^{24}
Ti	10^{21}
Pu	10^{20}
W	10^8
Tc	10^3
Co	10^0

Source: Copeland et al. 1978

In another study, Ren et al. (1994) conducted a series of laboratory experiments to optimize the removal of uranium contamination from copper. Samples weighing 100 grams were doped with 238 ppm uranium and melted with various fluxes. The investigation showed that residual uranium in the copper was at a minimum when the basicity of the flux was about 1.1. The highest decontamination factors were obtained when the flux was made from a blast furnace slag with the nominal composition: 38.1% SiO_2 , 41.4 %CaO, 3.8 %MgO, 2.6% Fe_2O_3 , and 14.1% Al_2O_3 . To minimize the residual uranium in the copper, the mass of flux needed to be at least 5% of the metal charge. The researchers also found that over a range of uranium concentrations of 2.4 to 238 ppm, the residual uranium content in the copper ingot was unchanged. This is the opposite of the finding of Abe et al. (1985) discussed in the previous paragraph. The maximum decontamination factor achieved in the laboratory tests was 236.

Table C-14. Partitioning of Uranium in Laboratory Melts of Copper

Sample	Metal (g)	Flux (g)	U concentration (ppm)		Partition Ratio ^a	Flux Composition					
			Slag	Metal		Al ₂ O ₃	CaF	CaO	CuO	Fe ₂ O ₃	SiO ₂
1	100	10	934	0.13	718	25	—	25	—	—	50
2	100	10	341	0.37	92	20	—	20	—	—	60
3	100	10	411	0.11	374	15	—	15	—	—	70
4	100	10	213	0.14	152		—	30	5	—	65
5	100	10	265	0.54	49	10	—	20	5	—	65
6	100	10	390	0.45	87	10	—	30	5	—	55
7	100	10	1813	0.83	218	10	—	10	—	5	75
8	100	10	1273	0.04	3182	10	—	10	—	5	65
9	100	10	943	0.25	377	10	—	30	—	5	55
10	250	25	1590	1.36	117	borosilicate glass					
11	250	25	1650	0.14	1179	10	5	50	—	5	30
12	170	—	—	1.96	—	no flux					

Source: Heshmatpour and Copeland 1981

^a Mass of uranium in slag divided by mass in metal

Vorotnikov et al. (1969) studied the behavior of iridium and ruthenium during the electrorefining of copper. They used copper anodes with 0.4% Ni, to which Ru-106 and Ir-192 were added. The distribution of these radionuclides during electrorefining in laboratory cells at current densities of 175 to 350 A/m² is summarized in Table C-15.

Table C-15. Distribution of Iridium and Ruthenium During Electrorefining of Copper

Current Density (A/m ²)	Ir (%)			Ru (%)		
	Electrolyte	Slimes	Cathode	Electrolyte	Slimes	Cathode
175	14	84	none	65	29.8	3.8
240	15	83	none	67	27.4	3.2
350	15.5	81	none	70	20.1	3.0

Source: Vorotnikov et al. 1969

As can be seen, most of the iridium reports to the slimes, while most of the ruthenium reports to the electrolyte. The electrolyte was then decoppered at a current density of 400 A/m²; the

resultant solution was boiled to produce nickel sulfate. Distribution of the iridium and ruthenium after electrolyte purification is shown in Table C-16.

Table C-16. Distribution of Iridium and Ruthenium after Electrolyte Purification

Product	Ir (%)	Ru (%)
Regenerated Copper	None	5.0
Copper Sponge	Undetermined	21.0
Nickel Sulfate	Undetermined	12
Electrolyte	90	70

Source: Vorotnikov et al. 1969

Even after purification of the electrolyte, most of the iridium and ruthenium remain in that process stream.

C.4.1.3 Proposed Partitioning of Contaminants

Blast Furnace Smelting

Based on the information presented in Table C-5, expected partition ratios of contaminants during the processing of low-grade copper scrap in a blast furnace were developed using the studies of Opie et al. (1985) and Nelmes (1984). The study of Kusik and Kenahan (1978), also included in Table C-5, was not used to estimate partition ratios since those authors did not include information on slag compositions. The slag resulting from the blast furnace operation characterized by Opie et al. (1985) in Table C-5 is rich in recoverable metals. These authors describe a processing step in which the blast furnace slag is further treated in a EAF, to which 2% coke is added as a reductant (see Section C.2.3.1, Table C-6). The slag from this step is assumed to be granulated and sold. Slags generated from downstream operations are returned to the blast furnace for recovery of additional metal values. By assuming that the metal streams and the dust streams are combined, overall observed partitioning from the blast furnace/EAF processing can be calculated from the Opie study. This additional step was not used in analyzing the Nelmes data. The results of the partitioning studies are summarized in Table C-17. In developing this table, it was assumed that each 100 tons charged to a blast furnace produces 40 tons of black copper, 40 tons of slag, and 5 tons of baghouse dust (Nelmes 1984). To develop the ranges shown in Table C-17, the maximum and minimum values were selected from among the data from the various studies.

U.S. Patent No. 4,351,705 (related to the work of Opie et al. [1985]) provides information on the partitioning of silver. In one example from the patent, 1,455 tons of converter slag containing 17.2 oz/ton Ag were smelted in a blast furnace to produce 420 tons of black copper containing 43.2 oz/ton Ag and an unspecified quantity of blast furnace slag containing 0.81 oz/ton Ag. When the blast furnace slag was cleaned in an arc furnace, the silver content was reduced to 0.5 oz/ton. Based on additional information included in the patent, it can be estimated that approximately 1,170 tons of blast furnace slag were produced. The silver input to the smelting process from the converter slag was 25,000 oz; the silver output was 18,100 oz to the black copper and 950 oz to the blast furnace slag, leaving about 6,000 oz unaccounted for. In order to achieve a material balance, it is assumed here that the unaccounted material is contained in the baghouse dust. Using methodology similar to that for other metals during the slag cleaning process, one can estimate that the 950 oz of silver in the blast furnace slag are distributed as follows:

- black copper from EAF 410 oz
- slag from EAF 540 oz
- baghouse dust from EAF: set to zero (the quantity will be small relative to that collected in the converter baghouse).

These calculations provide the basis for the silver partition fractions in Table C-17.

Table C-17
Observed Partition Fractions in the Melting of Low-grade Copper Scrap in a Blast Furnace

Element	Metal		Dust		Slag	
	Min.	Max.	Min.	Max.	Min.	Max.
Cu	0.99	0.99	0.0023	0.0039	0.0027	0.011
Ni	0.73	0.97	0.0020	0.0053	0.023	0.27
Sb	0.80	0.84	0.056	0.060	0.10	0.14
Sn	0.89	0.91	0.028	0.066	0.019	0.068
Fe	0.14	0.24	0.00	0.00029	0.84	0.86
Zn	0.24	0.40	0.51	0.52	0.080	0.24
Pb	0.47	0.62	0.29	0.31	0.093	0.13
Cl	0	0	1.0	1.0	0	0
F	0	0	1.0	1.0	0	0
Ag	0.74	0.74	0.022	0.022	0.24	0.24

The observed partitioning during the smelting of copper scrap in a blast furnace, as summarized in Table C-17, is combined with chemical analogies for certain elements and thermodynamic predictions from Table C-12 to arrive at the proposed partitioning for the desired suite of elements. This summary is presented in Table C-18. Most of the actinides form very stable oxides and are expected to be removed from the copper and concentrated in the slag. Even if removal is not 100%, as proposed in Table C-18, when the black copper is blown in a converter, the strongly oxidizing conditions can be expected to remove residual quantities of these elements to the converter slag, which is recycled to the blast furnace.

Table C-18
Partition Fractions of Impurities in the Melting of Low-grade Copper Scrap in a Blast Furnace

Element	Metal	Slag	Baghouse Dust	Basis for Estimate
Ag	0.74	0.02	0.24	Table C-17
Am		1.0		Table C-12
Ce		1.0		Table C-12
Co	0.73/0.97	0.023/0.27	0.0020/0.0053	Same as Ni, Table C-13
Cu	0.99/0.99	0.0027/0.011	0.0023/0.0039	Table C-17
Cs		0.10/0.20	0.80/0.90	Table C-12, WCT
Fe	0.14/ 0.24	0.84/0.86	0.00/0.00029	Table C-17
Mn	0.14/0.24	0.84/0.86	0.00/0.00029	Same as Fe
Ni	0.73/0.97	0.023/0.27	0.0020/0.0053	Table C-17
Np		1.0		Table C-12, Table C-13
Pa		1.0		Table C-12
Pb	0.47/0.62	0.093/0.13	0.29/0.31	Table C-17
Pu		1.0		Table C-12, Table C-13
Ra		1.0		Table C-12
Ru	0.99/0.99	0.0027/0.011	0.0023/0.0039	Same as Cu
Sb	0.80/0.84	0.10/0.14	0.056/0.060	Table C-17
Si		some	some	Table C-5
Sr		1.0		Table C-12
Tc	0.73/0.97	0.023/0.27	0.0020/0.0053	Same as Ni, Table C-13
Th		1.0		Table C-12, Table C-13
U		1.0		Table C-12, Table C-13
Zn	0.24/0.40	0.080/0.24	0.51/0.52	Table C-17

WCT = Author judgement

Converting

Some information on the composition of the process streams emanating from a copper converter is presented in Table C-8. However, no mass balance information was available to develop estimates of partition ratios. If copper scrap is introduced directly into the converter, it is expected that partitioning will be similar to that in the blast furnace. The strongly oxidizing conditions should insure that any actinides and other strong oxide formers will be oxidized and removed with the slag. If the scrap were introduced at the blast furnace stage, removal of additional Fe, Ni, Sb, Sn, Pb and Zn would be expected, based on the information included in Tables C-5 and C-8, resulting in blister copper with fewer impurities.

Fire Refining and Electrolysis

Expected partitioning of impurities in fire-refined copper and in electrorefined copper is summarized in Tables C-19 and C-21, respectively. Both fire-refined copper and electrorefined copper are included since both are used to produce end products. For example, fire-refined copper is used to produce sheet and tubing while electrorefined copper is used to produce wire. The elemental partitioning proposed in Table C-19 is appropriate for evaluating scenarios involving production for non-electrical applications where, say, No. 1 scrap is used to make a copper product such as tubing for plumbing applications or sheet for roofing. If the scrap is introduced earlier in the process then, with the exception of silver and ruthenium, which are not easily oxidized, the quantities of radioactive contaminants remaining with the metal should have been reduced during prior processing steps. The values for Ag, Fe, Ni, Pb, Sb, and Zn were developed using the data in Table C-8 for the feed composition and the data of Garbay and Chapuis (1991) is cited in Table C-9 for the chemistry of the fire-refined anodes. While the use of two unrelated data sets is a recognized problem, better data were not uncovered during the current study. This concern is ameliorated, in part, by providing a range for many of the partition factors.

As was discussed in Section C.2.3.1, a reverberatory furnace used for fire refining may not be equipped with a baghouse for dust collection. Offgas exiting the furnace after-burner may be exhausted directly through a stack. There are no NESHAPS standards for secondary copper smelters.

Brunson and Stone (1975) provide information of the composition of the anode and cathode copper, as well as anode slimes at the Southwire Co. The compositions are listed in Table C-20.

Table C-19. Partition Fractions of Impurities in the Fire Refining of Copper

Element	Metal	Slag	Offgas	Basis for Estimate
Ag	0.30/0.59	0.41/0.70		Table C-8, Table C-12, Garbay and Chapuis 1991
Am	0.001/0.01	0.99/0.999		Same as Pu
Co	0.05/0.10	0.90/0.95		Table C-12, same as Ni
Cs		0.10/0.20	0.80/0.90	Table C-12, WCT
Fe	0.02/0.05	0.95/0.98		Table C-8, Table C-12, Garbay and Chapuis 1991
Mn	0.02/0.05	0.95/0.98		Table C-12, Same as Fe
Ni	0.05/0.10	0.90/0.95		Table C-8, Table C-12, Garbay and Chapuis 1991
Np	0.001/0.01	0.99/0.999		Same as Pu
Pa	0.001/0.02	0.98/0.999		Same as U
Pb	0.22	0.73/0.78	0.00/0.05	Table C-8, Table C-12, Garbay and Chapuis 1991, WCT
Pu	0.001/0.01	0.99/0.999		Tables C-12 and C-13, Heshmatpour et al. 1983
Ru	1			Table C-12
Sb	0.08/0.25	0.75/0.92	0.00/0.05	Table C-8, Table C-12, Garbay and Chapuis 1991, WCT
Si		1		Table C-12
Sr		1		Table C-12
Tc	0.001	0.999		Table C-12 and C-13
Th	0.001/0.02	0.98/0.999		Same as U
U	0.001/0.02	0.98/0.999		Tables C-12 and C-13, Heshmatpour and Copeland 1981 (Table C-14)
Zn	0.10/0.20	0.80/0.90	0.00/0.05	Table C-8, Table C-12, WCT, Garbay and Chapuis 1991

WCT = author judgement

Table C-21 presents partition fractions of selected impurities in the electrorefining process, based on the data reported by Brunson and Stone (1975). Cobalt and manganese were assumed to behave like nickel and iron, respectively. Strontium was assumed to behave similarly to calcium. When a contaminant was identified in both the anode slimes and in the cell bleed (i.e., Fe, Sb, and Zn), the unaccounted for material was assumed to accumulate in the nickel sulfate, which is recrystallized from the cell bleed after copper is removed in the liberator cells. Detailed calculations are summarized in Appendix C-1. Ruthenium partitioning is based on data of Vorotnikov et al. (1969). Metal partitioning can also be estimated for a limited suite of elements using the data of Ramachandran and Wildman (1987) presented in Section C.2.3.4. Comparing these data with the values in Table C-21 indicates that the latter values are conservative (i.e., show slightly higher partitioning to the metal) for use in predicting radiation exposures to residual radioactive contaminants in metal.

Table C-20
Composition of Anode and Cathode Copper and Anode Slimes at the Southwire Co.

Element	Typical Anode (%)	Typical Cathode	Anode Slimes (%)
Cu	99.50	99.99%	8.77
O	0.10	—	—
S	0.003	—	—
Pb	0.19	5 ppm	31.45
Ni	0.10	7 ppm	
As	0.005	1 ppm	0.75
Sb	0.010	1 ppm	—
Bi	0.0007	0.1 ppm	—
Au	0.0012	—	0.55
Ag	0.024	10 ppm	4.65
Se	0.031	0.5 ppm	—
Te	0.0003	1 ppm	—
Sn	0.025	1 ppm	9.28
Fe	0.025	6 ppm	1.20
Zn	0.013	—	—
Ca	—	—	1.10
Si	—	—	3.50

Source: Brunson and Stone (1975)

Note: Slimes also contain 0.001% Pt and 0.001% Pd.

The literature on the electrorefining of copper abounds with consideration of the removal of impurities typically associated with copper, including Ag, As, Bi, Ni, Pb, Sb, Se, and Te. Virtually no information was uncovered in the course of this study on actinides and fission products, which are among the possible contaminants of copper cleared from nuclear facilities. To provide a quantitative perspective on the expected behavior of these contaminants during electrorefining, recourse was taken to some general electrorefining principles. According to Demaeral (1987):

During the electrorefining of copper, anode impurities either dissolve in the electrolyte or remain as insoluble compounds in the anode slime. Elements less noble than copper such as zinc, nickel and iron easily dissolve in the electrolyte. Elements more electropositive than copper, e.g. selenium, tellurium, silver, gold, and the platinum group metals and elements which are insoluble in sulphuric acid, such as lead, are concentrated in the anode slime. A

third group of elements, comprising the impurities which have a dissolution potential comparable to copper, such as arsenic, antimony, and bismuth, behave in a different way. Depending on anode composition and other operational parameters they either report to the slime or to the electrolyte with a widely fluctuating distribution pattern. Further, these elements can, depending on the respective concentration in the electrolyte, undergo several side reactions in the bulk of the electrolyte, resulting in a wide range of insoluble compounds and floating slimes.

Table C-21. Partition Fractions of Impurities in the Electrefining of Copper

Element	Metal	Anode Slimes	Electrolyte Bleed
Ag	0.04	0.96	
Am			1.0
Ca		0.5	0.5
Co	0.01		0.99
Cs			1.0
Fe	0.02	0.36	0.62
Mn	0.02	0.36	0.62
Ni	0.01		0.99
Np			1.0
Pb	0.003	0.997	
Pu			1.0
Ru	0.03/0.04	0.65/0.70	0.20/0.30
Sb	0.01		0.99
Si		1.0	
Sn	0.001	0.999	
Sr		0.5	0.5
Tc			1.0
Th			1.0
U			1.0
Zn			1.0

Electrode potentials for half-cells of various elements less noble than copper are listed in Table C-22. From this tabulation, it can be deduced that all the listed elements should report to the electrolyte and that a fraction should be continuously removed from the electrefining circuit with the electrolyte bleed. In the absence of modifying information, all the elements less noble than copper are assumed to report 100% to the electrolyte. During treatment of the electrolyte

bleed, it is not known whether many of these elements would concentrate in the black acid or in the crystallized nickel sulphate. Based on its electrode potential, strontium is expected to concentrate in the electrolyte. However, as noted by Brunson and Stone (1975), some calcium (and, by chemical analogy, strontium) is found in the slimes. Since the calcium content of the anodes is not reported by these authors, a partition ratio cannot be calculated. For Table C-21 it was arbitrarily assumed that calcium (and strontium) is distributed equally between the electrolyte and the slimes. Most of the nickel and probably the zinc, iron, cobalt, and manganese would be recovered from the electrolyte bleed as mixed sulfate crystals¹³.

Table C-22. Half-cell Electrode Potentials of Elements less Noble than Copper

Reaction	Potential (V)
$\text{Cs} = \text{Cs}^+ + \text{e}^-$	-2.92
$\text{Sr} = \text{Sr}^{2+} + 2\text{e}^-$	-2.89
$\text{Am} = \text{Am}^{3+} + 3\text{e}^-$	-2.32
$\text{Pu} = \text{Pu}^{3+} + 3\text{e}^-$	-2.07
$\text{Th} = \text{Th}^{4+} + 4\text{e}^-$	-1.90
$\text{Np} = \text{Np}^{3+} + 3\text{e}^-$	-1.86
$\text{U} = \text{U}^{3+} + 3\text{e}^-$	-1.80
$\text{Zn} = \text{Zn}^{2+} + 2\text{e}^-$	-0.763
$\text{Tc} = \text{Tc}^{\text{x}+} + \text{x}\text{e}^-$	-0.71
$\text{Fe} = \text{Fe}^{2+} + 2\text{e}^-$	-0.44
$\text{Co} = \text{Co}^{2+} + 2\text{e}^-$	-0.277
$\text{Ni} = \text{Ni}^{2+} + 2\text{e}^-$	-0.25
$\text{Cu} = \text{Cu}^{2+} + 2\text{e}^-$	0.337

Sources: Lewis and Randall 1961, Snyder et al. 1987. (All values quoted by Snyder et al. (1987), except the one for Tc, were taken from Latimer 1953.)

Note: Potentials at 25°C

For copper wire and other electrical conductors produced from fire-refined copper, estimating the partition fractions of contaminants in the metal involves combining the factors in Tables C-19 and C-21. Thus, if there were 1 kg of lead in a unit of copper scrap, there would be 220 g of lead in the fire-refined copper and 0.7 g in the electrolytic copper.

¹³ Dobner (1997) has indicated that the composition of crude nickel sulfate ($\text{NiSO}_4 \cdot 2\text{H}_2\text{O}$) is 27% Ni, 0.7% Zn, 0.3% Fe, 0.18% As, and 0.12% Sb.

C.4.2 Partitioning During Brass Smelting

Partitioning of contaminants during brass smelting is expected to be different from that in fire refining of copper. In fire-refining operations, the objective is to remove, by oxidation and slagging, as many impurities as possible. In brass melting, on the other hand, one objective is to minimize losses of alloying elements such as Zn, Fe, Mn, Pb, Al, and Sn. Consequently, from a conservative perspective in assessing radiation exposures to radioactive contaminants in metal, it should be assumed that all the contaminants remain in the metal.

C.5 EXPOSURE SCENARIOS

C.5.1 Modeling Parameters

As discussed in the previous sections, there are numerous options for the introduction of copper scrap into the copper refining process. Worker exposures to the contaminated scrap prior to smelting would be relatively independent of where the scrap is introduced into the secondary recovery process but would vary with the type of scrap. Typical operations may involve sorting, shredding, briquetting, and transportation. Insulation removal is required for the recycling of most copper wire.

It is likely that slag generated at any step in the process will be returned to a blast furnace for further processing and only blast furnace (or cleaned blast furnace) slag will exit the process. This slag will be sold or disposed of. The blast furnace operation may be at a different location than the initial secondary smelting operation. In that case, haulage of contaminated slag may be required. Since slag volumes will be smallest when introducing No. 1 copper scrap directly into a fire-refining furnace, the concentrations of any radionuclides that partition to the slag will be greatest for that type of operation. This slag will be diluted when reprocessed in a blast furnace.

Scrap copper released from nuclear installations is likely to be carefully sorted high-quality material. As such, it would most likely be introduced into the secondary refining process at the fire refining stage where it would be used to produce anodes for electrorefining or finished mill products such as sheet and tubing. Expected partitioning of contaminants during fire refining is summarized in Table C-19. While additional partitioning occurs during electrorefining, the result of that process is to further reduce the impurities in the metal. Therefore, it is unlikely that electrorefining of cleared scrap would lead to higher radiation exposures than received during the

fire-refining of such scrap. Possible exceptions could be exposures to anode slimes and electrolyte bleed streams from the electrolysis cells.

C.5.1.1 Dilution of Cleared Scrap

The information presented in Section C.1.1 indicates that a maximum of 10,833 t of copper scrap would be cleared in any one year. This represents about 0.8% of the total annual consumption of copper scrap, as listed in Table C-4. Thus, if this scrap were uniformly distributed amongst all consumers, the dilution factor would be 0.008. If all this scrap was processed through a single 200-ton reverberatory furnace, which has an annual capacity of 45,500 tons (~41,300 t) the dilution factor would be 0.26. This calculation assumes that the furnace operates 330 days per year on a 24-hour cycle with 25% of the charge left in the furnace to facilitate the subsequent melting cycle. A more reasonable assumption is that the reference facility—the 200-ton reverberatory furnace cited above—would process the 2,080 t/a of copper scrap generated during the decommissioning of the K-25 Plant at Oak Ridge, while the scrap stockpiled during the years when no scrap was cleared by DOE would have a different disposition. In such a case, the dilution would be 0.05.

C.5.1.2 Slag Production

Slag production in a reverberatory furnace varies as a function of the percentage of copper in the charge. With increasing copper grade (Biswas and Davenport 1976):

- Copper concentration in slag increases
- Slag weight decreases
- Copper loss decreases

High-copper-content scrap metal, ranging from 85-95% copper, loaded in a 350-ton-per-day reverberatory furnace, may generate about 30 tons per day of slag. The slag contains an economically recoverable concentration of copper, which may be recycled to a blast furnace for recovery (Murrah 1997). Slag is used for the manufacture of abrasives, shingles, road surface bedding, mineral wool, and cement/concrete materials (Carey 1997).

Slags from a Peirce-Smith converter have an economically viable copper content and may be recycled to a reverberatory or blast furnace to reduce copper loss (Biswas and Davenport 1976).

The process options are myriad; each processor has its own preferred operational cycle. These range from simple remelting and casting, to smelting and recycling the slag, depending upon the available options (Murrah 1997).

One producer, who uses a reverberatory furnace to melt high grade copper scrap and cast logs from which extrusion billets are cut, estimates that the slag weight is about 2 to 2.5% of the charge weight (Burg 1999).

Based on the available information, it is proposed for modeling purposes that a reverberatory furnace melting and fire refining No. 1 copper scrap generates 0.02 tons of slag per ton of scrap charged. Since many oxidizable impurities concentrate in the slag, a small slag volume will increase concentrations of these elements in the slag.

C.5.1.3 Baghouse Dusts

In the copper conversion process, baghouse filtration is used at various processing stages to collect zinc, tin and lead dusts. The composition of the dust is a function of the copper charge composition. Thus, dust capture will vary strongly with alloy composition. Assuming a typical converter charge, about 0.25% of the copper in the feed will enter the baghouse collection system as oxide. Dust, depending on the alloy composition of the charge, is sent to lead, zinc, or tin smelters to recover these metals (Edelstein 1997).

In a reverberatory furnace, the dust produced may be as much as 1% of the charge. The dust is frequently recycled to the furnace if the copper content is significant. Dust from a Peirce-Smith converter may contain as much as 11% copper; it is almost always recycled to a smelting furnace (Biswas and Davenport 1976). The mass of dust generated by an EAF used for copper smelting is about 0.25% of the mass of scrap metal charged to the furnace.

However, as noted previously, some operations do not use a baghouse for dust control, so that the species that accumulate in the offgas, as noted in Table C-19, would be released to the atmosphere.

C.5.1.4 Electrolyte Bleed

During the final electrolytic purification of copper, part of the electrolyte is bled off to control impurity build-up in the electrolytic cells. The soluble impurities include As, Bi, Co, Fe, Ni, Sb, and Zn. As noted in Section C.4.1.3, As, Bi, and Sb may report either to the electrolyte or to the anode slimes depending on such factors as anode chemistry and cell operating parameters. Actinide elements are also assumed to report to the electrolyte. Some of these impurities are removed from the bleed stream by evaporation and crystallization and may be contained in products which are sold. Other impurities may remain in the electrolyte and be returned to the electrorefining process or used to leach slimes.

The implication is that this added step in the processing of copper creates the potential for a new source of exposure by reconcentrating residual metals. However, most of the residual radioactive contaminants in the cleared copper scrap will have partitioned to the slag or been removed in the offgas well before this stage. The principal exceptions are isotopes of Co, Fe, Ni, Ru, and Zn. If a large electrolytic refinery uses 460,000 tpy of copper anodes containing 0.1% Ni, the nickel content in the feed is 460 tons. According to Table C-21, 99% of Ni is concentrated in the electrolyte bleed stream. If this nickel is crystallized as NiSO_4 , which is 38% Ni by weight, and if the crude nickel sulfate contains 5% H_2SO_4 and 3% water, then the annual production of the crude precipitate is about 1,300 tons ($460 \times 0.99 \div [0.92 \times 0.38] \approx 1,300$). The concentration of nickel in the crude nickel sulfate is 35% ($0.38 \times 0.92 = 0.35$), or about 350 times that of the nickel in the anodes. By chemical analogy, cobalt should be similarly concentrated. While the behavior of other impurities in the electrolyte bleed is unknown, it is likely that some of these will be crystallized with the nickel sulfate.

According to Garbay and Chapuis (1991), a 50,000-t French electrorefining plant produces about 500 t of residual sulfuric acid, about 30 t of arsenical sludge, and about 60 t of nickel sulfate. The nickel sulfate production rate quoted by Garbay and Chapuis—1.2 kg/t of Cu—is lower than that described in the previous paragraph—equivalent to 2.9 kg/t of Cu—partly because the nickel content in the French anodes is only 0.05% (see Section C.2.3.3).

C.5.1.5 Anode Slimes

Brunson and Stone (1975) cite a slimes generation rate of 15 lb of anode slimes produced per ton of copper refined at the Southwire Co. This rate of slimes production—7.5 kg/t of Cu—is more than an order of magnitude higher than the 600 g/t quoted by Garbay and Chapuis (1991). The

cause of this difference is not known. However, data quoted by Schloen (1987) corresponds to slimes generation rates ranging from 1 to 7.3 kg/t of anodes for nine U.S. electrolytic refineries, suggesting that the higher figure is more typical of U.S. experience.

C.5.1.6 Summary Model for Fire-Refined Products

Based on the information presented above, the following model is proposed for fire-refined products, such as copper tubing.

A 200-ton reverberatory furnace is used to melt No. 1 copper scrap. The furnace operates 12 out of every 14 days, with two days down for routine maintenance. The furnace also is shut down for an additional two weeks per year for major maintenance. The furnace operates on a 24-hour cycle with the following cycle elements :

- Charging 4.5 hr
- Melting 4.5 hr
- Refining and slagging 5.5 hr
- Poling 2.5 hr
- Casting 7 hr

Since about 25% of the melt remains in the furnace as a heel for the subsequent heat, the daily output is 150 tons and the annual output is 45,000 tons. The annual furnace input is 45,500 tons of copper scrap. The furnace produces 910 tons of slag and 110 tons of dust (dust generation of about 5 lb per ton) annually. The slag contains about 40% copper and the dust contains about 75% copper. The dust is either collected in the baghouse or released to the atmosphere. The slag and the dust (if captured) are sent to an outside processor for recovery of additional metal values. Elemental partitioning is presented in Table C-19. The approximate material balance is illustrated in Figure C-4.

The slag from the reverberatory furnace is shipped to an outside processor who treats the material in a 50 tph blast furnace with an annual capacity of 36,000 tons ($50 \text{ tph} \times 24 \text{ hr/day} \times 300 \text{ days/year} = 36,000 \text{ tons}$). Thus, the slag from the reverberatory furnace undergoes a further dilution of 0.025 ($910 \div 36,000 \approx 0.025$). The blast furnace slag is then sold for industrial applications such as use in abrasives, roofing materials, or road building materials.

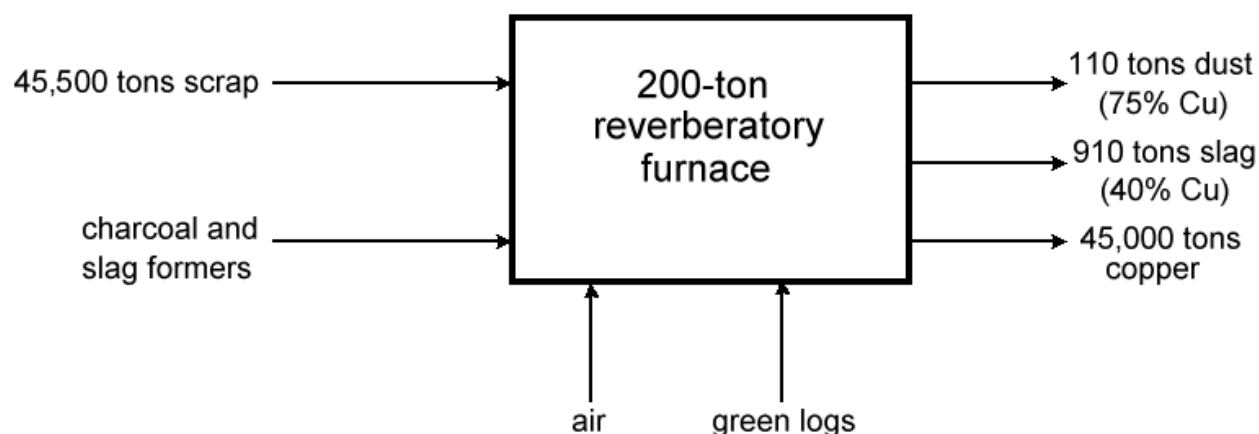


Figure C-4. Proposed Material Balance for Modeling Copper Produced by Fire Refining (values are rounded)

C.5.1.7 Summary Model for Electrorefining

Based on the previously presented information, the following model is proposed for high conductivity electrical products, such as wire and cable, which require electrorefining after fire refining for further impurity removal.

Annual output from the electrolytic refinery is 450,000 tons of copper, 3,200 tons of anode slimes, and 1,300 tons of crude nickel sulfate (Schloen 1987). Sulfuric acid recovered from the electrolyte bleed circuit is assumed to be used for electrolyte makeup; accordingly, it is returned to the process. The nickel sulfate, containing 5% H_2SO_4 and 3% H_2O , is sold to nickel producers for metal recovery. The nickel sulfate also contains contaminants, such as iron and zinc.

The annual input to the reverberatory furnace at the electrolytic refinery is assumed to be 24,000 tons of No. 2 copper scrap and 102,000 tons of blister copper from primary producers. The average nickel content of the anodes is 0.1%.

An approximate material balance is presented in Figure C-5. Elemental partitioning can be calculated by combining the factors included in Tables C-19 and C-21.

C.5.2 Worker Exposures

Dust sampling at a primary copper smelter has been reported by Michaud et al. (1996). Samples were taken at a smelting furnace and a converter located in separate buildings. Results are

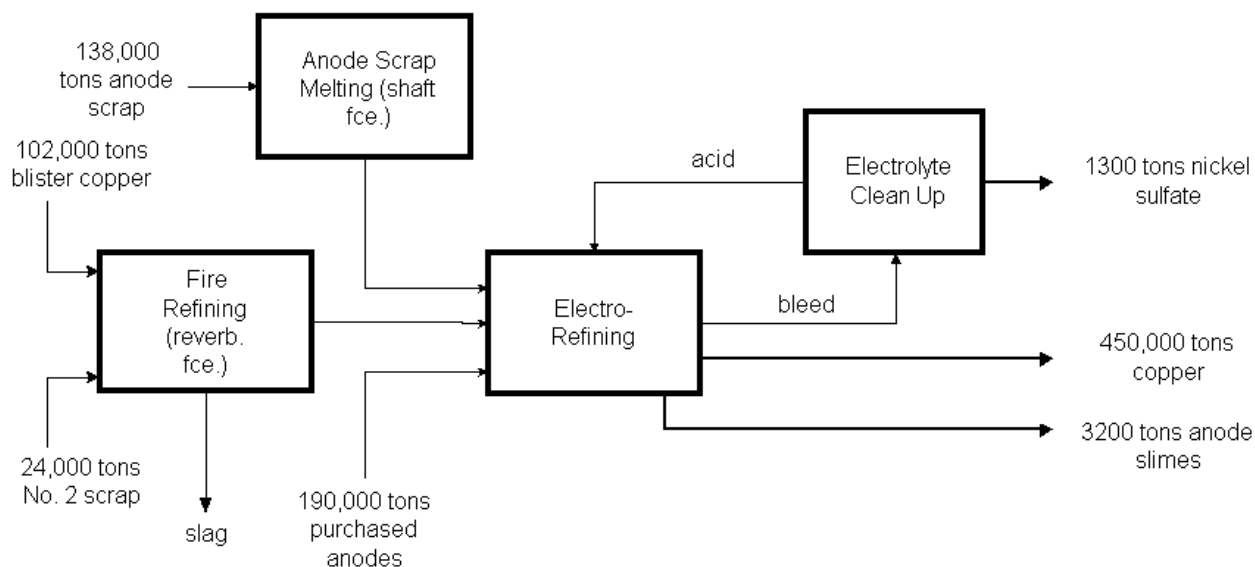


Figure C-5. Simplified Material Balance for Electrorefining of Copper Produced from Scrap

summarized in Table C-23. Cadmium and nickel were not detected in the dusts.

Table C-23. Airborne Dust Concentrations At Primary Copper Smelter (mg/m³)

Unit	Total	Respirable	Lead	Copper	Arsenic
Smelting Furnace	2.3	0.6	0.21	0.10	0.02
Converter	2.1	0.8	0.15	0.32	0.02

Source: Michaud et al. 1996

C.5.2.1 Baghouse Dust Agglomeration Operator

As noted in Table C-19, cesium is the main contaminant that would distribute to the offgas during fire refining of copper scrap. The exposure scenario developed here is designed to capture worker exposure to this dust and is based primarily on information presented in Section C.2.3.8.

Basic assumptions include:

- Copper output 342,000 tpy
- Baghouse dust from fire-refining furnaces 51,100 tpy
- Cesium partitioning to dust 90%

Based on these assumptions, the dust generation rate will be 0.15 tons of dust per ton of copper product ($51,100 \div 342,000$). The cesium reconcentration factor due to preferential partitioning to the dust will be 6:1 ($5,000 \times 0.9 \div 750$). The operator would be exposed for 7 hours per day, 5 days per week to the mass of wetted dust in a concrete bunker that is about $20 \times 30 \times 12$ ft high. It is assumed that the bunker contains a maximum of three days' output from the agglomerator or 420 tons ($20 \text{ tph} \times 7 \text{ hr/d} \times 3 \text{ d} = 420 \text{ tons}$).

If the recycling facility used a reverberatory furnace without a baghouse, then all the cesium would be exhausted up the stack and become airborne.

C.5.2.2 Furnace Operator

A furnace operator would be part of a crew that spends full time in the vicinity of the reverberatory furnace that holds 200 tons of copper. For about two hours per shift, he would be standing 5 to 10 ft from an open furnace, skimming slag from the furnace with a rake into a metal box about $4 \times 4 \times 1$ ft. Another operator would transport the slag box with a forklift truck about 200 ft to an area on the furnace room floor where the box is dumped. The cooled slag is broken up by an operator with a pneumatic hammer; copper is then culled by hand from the slag. At other times the operator will be shoveling charcoal and slag-forming agents into the furnace or tapping the furnace to allow the molten metal to flow through launders to the holding furnace.

C.5.2.3 Scrap Handler

The scrap handler would spend full time in the vicinity of the scrap piles preparing the material for charging into the furnace. This might include loading material into a briquetting machine and transporting the briquetted scrap to a staging area with a fork-lift truck. On average, about 200 tons of scrap are stockpiled in the scrap-handling area.

C.5.2.4 Casting Machine Operator

A casting machine operator would cast the copper into logs and assist in moving the cooled logs from the casting machine cooling pit to the billet-cutting machine. The operator would spent full time working near several copper logs that are about 26 feet long and up to 12 inches in diameter.

C.5.2.5 Scrap Metal Transporter

If all the scrap from the largest annual DOE source (i.e. 2,080 t from the K-25 plant in Oak Ridge) were shipped to Southwire in Carrollton, Ga. for recycling, 104 shipments in a 20-t truck would be required. The distance is about 250 miles; the estimated driving time is six hours. Thus the total driver exposure would be about 624 hours. Other situations, which would lead to greater exposures, are possible. To accommodate this possibility, it is conservatively assumed that a truck driver spends full time driving a 20-t truck, with the truck loaded only one-half of the time (i.e., about 1,000 hr/y).

C.5.2.6 Tank House Operator

A tank house operator in a 450,000 tpy electrolytic refining plant would collect and drum 3,200 tons of anode slimes for transport to a refinery for metals recovery.

C.5.3 Non-Industrial Exposures

C.5.3.1 Driver of Motor Vehicle

The average amount of copper used in automobiles or light trucks is 50 pounds. The radiator contains about 80% of this; the electrical system contains about 20%. These elements are mostly under the hood presenting minimal exposure hazards. The radiator would consist of recycled scrap (CDA 1997). It is likely that the copper would come from several lots of material with differing processing histories.

C.5.3.2 Homemaker

Home appliances and heating and cooling systems contain copper produced from recycled scrap. Copper usage in home appliances is as follows (CDA 1997):

- Central Air Conditioner 50 lb
- Refrigerator 5 lb
- Dishwasher 5 lb
- Washing Machine 4.4 lb
- Dryer 2 lb
- Range 1.3 lb

- Garbage Disposer 2.3 lb
- Dehumidifier 2.7 lb
- Heat Pump 48 lb

Radiation exposures from any residual radioactive contaminants in these products would be very low relative to those associated with handling copper scrap and finished and semi-finished products made from this metal during the various stages in the copper refining process. This is primarily because of the small quantities of copper in these products, and because the copper would be obtained from many different lots of material, not all of which would be produced from cleared scrap.

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APPENDIX C-1

PARTITIONING DURING FIRE REFINING AND ELECTROREFINING OF COPPER SCRAP

Table C1-1. Partitioning During Fire Refining and Electrolysis of Copper Scrap

Reverb charge	45500 tons														
Reverb output	910 tons in slag	910 tons at 40% Cu													
	110 tons in dust	110 tons at 75% Cu													
	45000 tons in anode Cu														
Electrolytic Cell output	44500 tons as cathodes														
	337.5 tons as slimes	15 lb/ton													
	128.7 tons as nickel sulfate (38%Ni)														
													Adjusted	Adjusted	Adjusted
	Anodes	Cathodes	Metal	Slimes	Slimes	Slimes	Bleed	Bleed	Material Balance	Slimes	Bleed	Metal	Partition		
	(wt. %)	tons	(ppm)**	tons	Partition	(wt %)	tons	Partition	tons	Partition	tons unaccounted	Partition	Partition	Partition	Check
Cu	99.5	44775	99.99%	44495.55		8.77	29.60								
Ni	0.1	45	7	0.31	0.0069	0	0.00	0.000	44.69	0.99	0.00		0.000	0.993	0.0069 1.0000
Sb	0.01	4.5	1	0.04	0.0099	0	0.00	0.000			4.46	add bal. to bleed	0.000	0.990	0.0099 1.0000
Sn	0.025	11.25	1	0.04	0.0040	9.28	31.32	2.784			-20.11	add bal. to anodes	0.999	0.000	0.0014 1.0000
Fe	0.025	11.25	6	0.27	0.0237	1.2	4.05	0.360			6.93	add bal. to bleed	0.360	0.616	0.0237 1.0000
Zn	0.013	5.85	0	0.00	0.0000	0	0.00	0.000			5.85	add bal. to bleed	0.000	1.000	0.0000 1.0000
Pb	0.19	85.5	5	0.22	0.0026	31.45	106.14	1.241			-20.87	subt. bal. fr. slimes	0.997	0.000	0.0026 1.0000
Ag	0.024	10.8	10	0.45	0.0412	5.2	17.55	1.625			-7.20	subt. bal. fr. slimes	0.959	0.000	0.0412 1.0000
Bi	0.0007	0.315	0.1	0.00	0.0141	0	0.00	0.000			0.31	add bal. to bleed	0.000	0.986	0.0141 1.0000
As	0.005	2.25	1	0.04	0.0198	0.75	2.53	1.125			-0.33	subt. bal. fr. slimes	0.980	0.000	0.0198 1.0000
Te	0.0003	0.135	1	0.04	0.3296	0	0.00	0.000			0.09	add bal. to slimes	0.670	0.000	0.3296 1.0000
Se	0.031	13.95	0.5	0.02	0.0016	0	0.00	0.000			13.93	add bal. to slimes	0.998	0.000	0.0016 1.0000
Ca						1.1	3.71	0.500*			-3.71	add bal. to anodes	0.500	0.500	0.0000 1.0000
Si						3.5	11.81	1.000*			-11.81	add bal. to anodes	1.000	0.000	0.0000 1.0000
Total		44965.8		44497			194.91				-32.46				
			**unless other units shown					* assumed							
							140 tons of slimes not accounted for								